TRANSLATION

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION	See Form PCT/IPEA/416
PCT03/1431		
International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/IB2004/000257	27.01.2004	
International Patent Classification (IPC) or national classification and IPC		
C07C 51/02, C07C63/26		
Applicant		
FREGOSO-INFANTE, Arturo, Guadalupe		

1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of _____ sheets, including this cover sheet.

3. This report is also accompanied by ANNEXES, comprising:

   a. ☒ *(sent to the applicant and to the International Bureau)* a total of 4 sheets, as follows:

      ☒ sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).

      ☐ sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.

   b. ☐ *(sent to the International Bureau only)* a total of (indicate type and number of electronic carrier(s))

      _____, containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).

4. This report contains indications relating to the following items:

☒	Box No. I	Basis of the report
☐	Box No. II	Priority
☐	Box No. III	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
☐	Box No. IV	Lack of unity of invention
☒	Box No. V	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
☐	Box No. VI	Certain documents cited
☐	Box No. VII	Certain defects in the international application
☐	Box No. VIII	Certain observations on the international application

Date of submission of the demand	Date of completion of this report
Name and mailing address of the IPEA/ES	Authorized officer
Facsimile No.	Telephone No.

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/IB2004/000257

Box No. I Basis of the report

1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ This report is based on translations from the original language into the following language _____, which is the language of a translation furnished for the purposes of:

☐ international search (Rule 12.3 and 23.1(b))

☐ publication of the international application (Rule 12.4)

☐ international preliminary examination (Rule 55.2 and/or 55.3)

2. With regard to the **elements** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

☐ the international application as originally filed/furnished

☒ the description:

pages 1-11 as originally filed/furnished

pages* _____ received by this Authority on _____

pages* _____ received by this Authority on _____

☒ the claims:

nos. _____ as originally filed/furnished

nos.* _____ as amended (together with any statement) under Article 19

nos.* _____ received by this Authority on _____

nos.* 12-15 received by this Authority on 06.01.2006

☐ the drawings:

sheets _____ as originally filed/furnished

sheets* _____ received by this Authority on _____

sheets* _____ received by this Authority on _____

☐ a sequence listing and/or any related table(s) – see Supplemental Box Relating to Sequence Listing.

3. ☒ The amendments have resulted in the cancellation of:

☐ the description, pages _____

☒ the claims, nos. 1-11 received by this Authority on 05.05.2005

☐ the drawings, sheets/figs _____

☐ the sequence listing *(specify)*: _____

☐ any table(s) related to sequence listing *(specify)*: _____

4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).

☐ the description, pages _____

☐ the claims, nos. _____

☐ the drawings, sheets/figs _____

☐ the sequence listing *(specify)*: _____

☐ any table(s) related to sequence listing *(specify)*: _____

* *If item 4 applies, some or all of those sheets may be marked "superseded."*

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/IB2004/000257

Box No. V **Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

Novelty (N)	Claims	1-17	YES
	Claims		NO
Inventive step (IS)	Claims	1-8	YES
	Claims	9-17	NO
Industrial applicability (IA)	Claims	1-17	YES
	Claims	1-17	NO

2. Citations and explanations (Rule 70.7)

Documents taken into consideration:

D1: GB 822 834 A (04/11/1959)

D2: Advances in Polymer Technology, 21 (4), pages 250-259 (2002)

D3: Polymer Journal, Vol. 29 (9), pages 708-712 (1997)

D4: US 6 580 005 B1 (17/06/2003)

The subject matter of the invention relates to a chemical method for efficiently recycling waste polyethylene terephthalate (PET), comprising (a) a first step of saponification (by a reaction with a strong base in an alcoholic medium and at the boiling point of said alcoholic medium) to give the terephthalic acid salt; (b) a step of separating out said salt; (c) a step of precipitating the terephthalic acid by means of a treatment with a strong acid; (d) a step of separating out said acid; and (e) a step of recovering the ethylene glycol from the alcoholic medium. There are two possible alternatives to this method, in which the solvent used is either a mixture of **non**-water-**miscible** alcohols (claim 1) or water-**miscible** alcohols (claim 9).

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

Document D1 describes a multiple-step method for recovering PET from waste materials by hydrolysis in an alcoholic medium (saponification), preferably ethanol or methanol, using an alkali metal hydroxide as the base (claims 1 and 6), followed by separation of the resulting salt and acidification thereof to give terephthalic acid (page 2, lines 41-44). The method is also useful for recovering the ethylene glycol generated during hydrolysis (page 1, lines 64-69). Indeed, examples 2 and 3 describe the hydrolysis of polyethylene terephthalate with two KOH equivalents, at the reflux temperature, under atmospheric pressure and for one hour, using 99.7 % methanol and 97.8 % ethanol, respectively, as the solvent. The precipitated potassium polyethylene terephthalate is filtered, dissolved in water and acidified to precipitate out the resulting terephthalic acid, which is isolated with a yield of 98 % (example 2) and 96 % (example 3).

Document D2 also describes a method for recycling PET from bottles and other used containers, and recovering terephthalic acid by alkaline hydrolysis in an alcoholic medium. Specifically, the reaction is carried out using potassium hydroxide as the base dissolved in 2-methoxy ethanol, under atmospheric pressure and at 120 °C, the reflux temperature of the solvent, for 2.5 hours (see pages 253-254 and 256). Once hydrolysis is complete, the reaction mixture is cooled and filtered. The resulting potassium terephthalate is dissolved in water and acidified with sulphuric acid to precipitate the terephthalic acid, which is then separated by filtration and washed with methanol. In these methods, the ethylene

glycol generated following hydrolysis of the polyethylene terephthalate is recovered by evaporation (page 252).

Document D3 describes a method carried out in an alcoholic reaction medium for simultaneously recovering terephthalic acid and ethylene glycol from PET from plastic waste by means of alkaline decomposition caused by a reaction with a stoichiometric amount of sodium or potassium hydroxide, in which the solvent is a mixture of an alcohol such as methanol or ethanol and an ether such as tetrahydrofuran, dioxane or dimethoxyethane. Hydrolysis is carried out at a temperature below 80 °C and once it is complete, the terephthalic acid salt is separated from the reaction medium by filtration, dissolved in water and acidified with hydrochloric acid. The resulting terephthalic acid is separated by filtration and the ethylene glycol is recovered from the filtrate by distillation (see pages 708-709; particularly the abstract).

Document D4 cited by the applicant describes a method (column 2, lines 12-38; column 4, lines 21-47) for recovering terephthalic acid from waste polyethylene terephthalate containers, which method comprises the same steps as the method of the present application, i.e. (1) subjecting the PET to a decomposition reaction in an alcoholic solvent such as ethylene glycol or 1,2-ethanediol, at a temperature of 120 to 190 °C (the boiling point of the solvent) and under atmospheric pressure (column 16, lines 12-16); (2) performing solid-liquid separation of the terephthalic acid salt and the solvent; (3) dissolving said salt in water; (4)

neutralising the salt; and (5) performing solid-liquid separation of the resulting terephthalic acid crystals.

The technical subject matter of claim **1** relates to the method for recycling PET characterised in that the first saponification step is carried out in a **non**-water-**miscible** reaction medium such as octanol, 1-pentanol and 1-pentanol/ethanol (examples 1, 4 and 7 of the application), whereafter the terephthalic acid salt is separated by adding water to the reaction medium to give two phases, namely an alcoholic phase containing the ethylene glycol and an aqueous phase in which the salt is dissolved and which is subjected to the acidification step (c) once it has been separated from said other phase.

None of documents D1 to D4 mentions the possibility of carrying out the method using a non-water-miscible alcoholic medium as the solvent. It follows that said documents do not disclose the invention defined in claim 1. Said documents likewise do not contain any suggestions that might lead a person skilled in the art to said method as an efficient process for recovering compounds that have a high market value.

As a result, the invention defined in claims **1 to 8** is considered to be novel, to involve an inventive step and to be industrially applicable as per the requirements of PCT Article 33(2), (3) and (4).

The technical subject matter of claim **9** relates to the method characterised in that the first saponification

step (a) is carried out in an alcoholic reaction medium consisting of a mixture of alcohols, and followed by step (b) in which the terephthalic acid salt is separated by filtration of the resulting crystals, which are then washed and treated with an acid as per step (c). Given that the terephthalic acid salt separation step is carried out by filtration of the crystals resulting from the saponification step, it can be deduced that said saponification step is carried out in a water-miscible medium, i.e. that the component alcohols of the mixture referred to in step (a) are water-**miscible** alcohols, as demonstrated in claim 11, which specifically mentions alcohols having 1 to 3 carbon atoms.

None of documents D1 to D4 mentions the use of mixtures of (water-miscible) alcohols as the solvent in PET recycling methods. It follows that the invention defined in claim 9 is novel over said documents. However, since said documents do describe the use of various water-miscible alcohols in various non-aqueous reaction media, alone or in combination with an ether component, and no technical advantage (higher yield, milder reaction conditions, etc.) is associated with the alternative embodiment as per the subject matter of claim 9, it is considered that the use of alcoholic media consisting of mixtures of said water-miscible alcohols would be obvious to a person skilled in the art, particularly because said use does not involve altering the operating conditions of the prior art PET recycling method.

It follows that the subject matter of claims **9 to 17** does not involve an inventive step in the light of documents

Box No. V **Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

D1 to D4 (PCT Article 33(3)).